EQL Memorandum No. 12

LEAD AS A TRACER FOR AUTOMOTIVE PARTICULATES: PROJECTING THE SULFATE AIR QUALITY IMPACT OF OXIDATION CATALYST-EQUIPPED CARS IN LOS ANGELES

by

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ABSTRACT

An analysis of the fate of lead in the Los Angeles Basin is used to evaluate an emissions to air quality model for automotive exhaust particulates. The dispersion model is then applied to projecting the annual average sulfate air quality impact of direct sulfuric acid mist emissions from oxidation catalyst-equipped cars of the 1975 model type. Estimates are given of the incremental sulfate contributions from three model years of oxidation catalyst-equipped cars burning a relatively low sulfur gasoline, and from roughly ten model years of 1975-type autos burning gasoline of sulfur content equal to that of the entire 1974 Southern California gasoline pool. In the latter case, sulfate concentrations in portions of downtown Los Angeles in 1985 could be elevated by roughly two thirds above present average sulfate values.

Introduction

In an effort to reduce automotive exhaust hydrocarbon and carbon monoxide emissions, nearly all 1975 model cars sold in California have been equipped with oxidation catalysts. But in addition to altering the gaseous composition of the vehicle exhaust, oxidation catalysts have the potential to change the character of the currently unregulated particulate emissions from automobiles. Trace amounts of sulfur present in gasoline may be oxidized by catalytic emission control systems to form particulate sulfates and sulfuric acid mist. Sulfates and sulfuric acid mist contribute to visibility reduction (White, et al., 1975) and are of current public health concern (National Research Council, 1975). Thus projection of the extent to which new catalyst technologies may affect sulfate air quality can be an important factor in deciding how to control emissions from future model automobiles.

The atmospheric aerosol measured at a given location reflects the combined particulate contributions of a variety of emission sources. Friedlander and his co-workers (Miller, et al., 1972; Friedlander, 1973; Gartrell and Friedlander, 1975) have shown that the characteristic chemical element signatures of specific emission sources can be used to trace the relative contribution of each source type to a set of aerosol samples. For example, material balance calculations performed by Huntzicker, et al.

(1975) indicate that virtually all of the aerosol lead present in the Los Angeles atmosphere can be attributed to the use of tetraethyl lead as an anti-knock compound in gasoline. By using suspended lead as a tracer for particulate emissions from the automobile, this paper will explore a simple dispersion model for automotive-related particulate air quality in the Los Angeles Basin. Having thus established the approximate emissions to air quality relationship for auto exhaust particulates, direct sulfate and sulfuric acid mist emissions from catalyst equipped cars can be examined for their possible long term impact on air quality.

Los Angeles Suspended Lead Air Quality

The modeling region chosen is a square, 80.52 km on each side, overlaying the central portion of the Los Angeles Basin as shown in Figure 1. For the purposes of this discussion automotive particulate emissions will be treated as arising from a grid of uniform area sources of strength proportional to local traffic density. Thus the region is further subdivided into a network of grid squares, each 3.22 km on a side, for which the 1969 spatial distribution of freeway and surface street traffic has been given by Roth, et al. (1974), as shown in Figures 2 and 3. Since we are interested only in lead from gasoline fueled vehicles, some adjustment to vehicle miles traveled must be made to reflect the use of diesel fuel and liquified petroleum gas. Thus 1969 auto traffic density used in this model will be reduced to 93.66 percent of the values shown in Figures 2 and 3

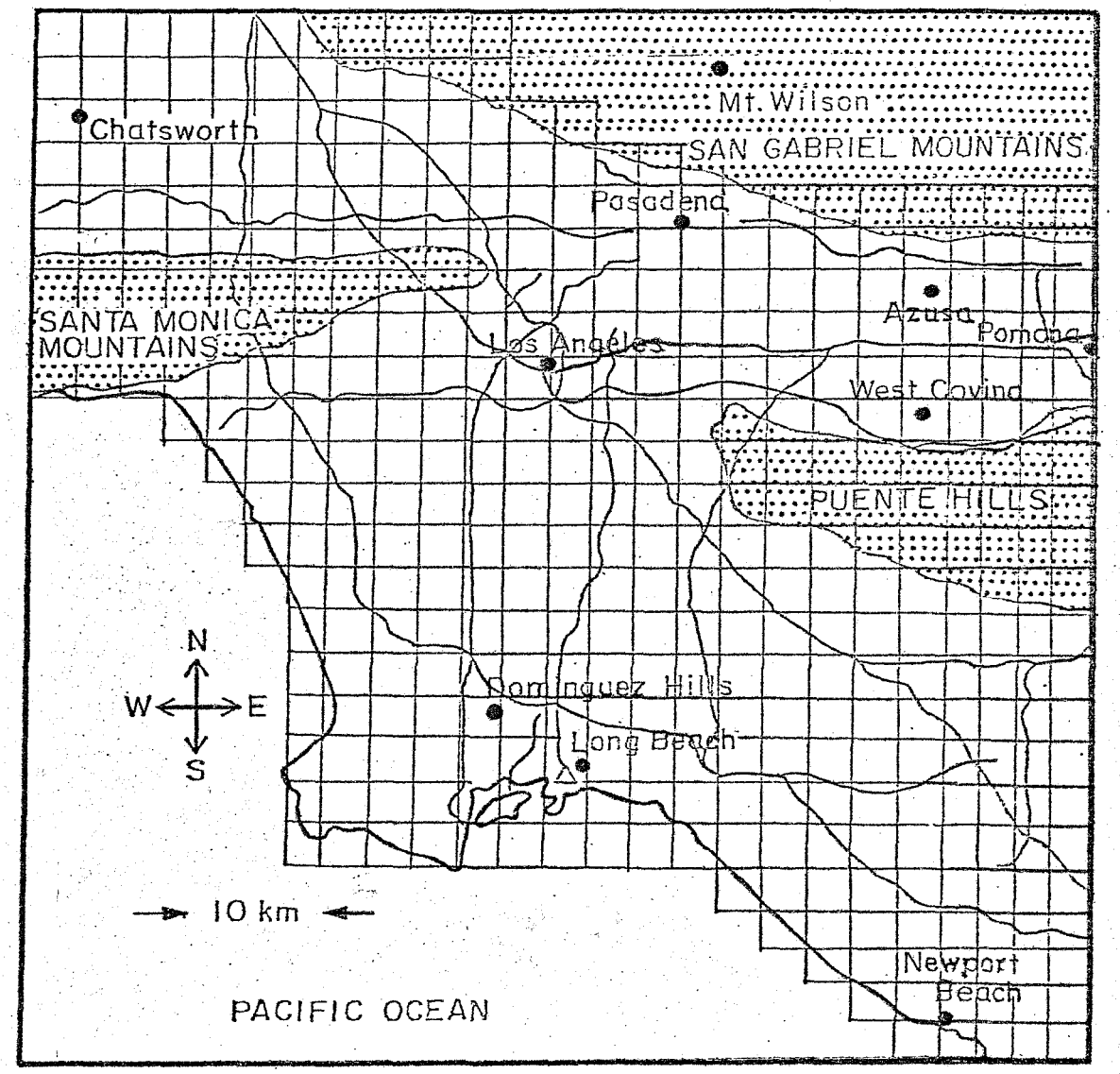

Figure 1 The Modeling Region (showing the grid network used and the Los Angeles freeway system in 1969).

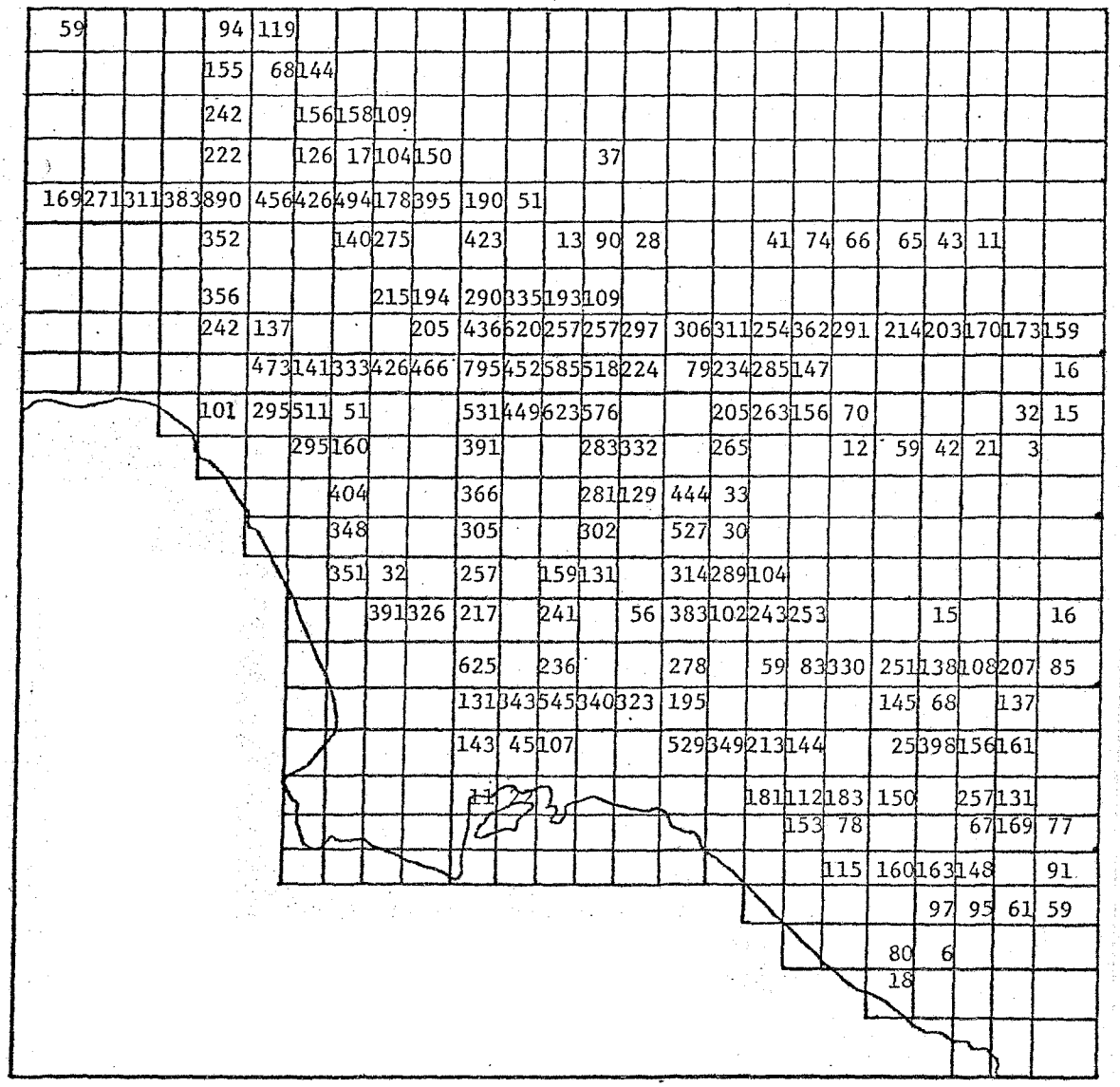

Figure 2 The Spatial Distribution of Freeway Traffic, 1969 (thousands of vehicle miles traveled daily within each grid square).

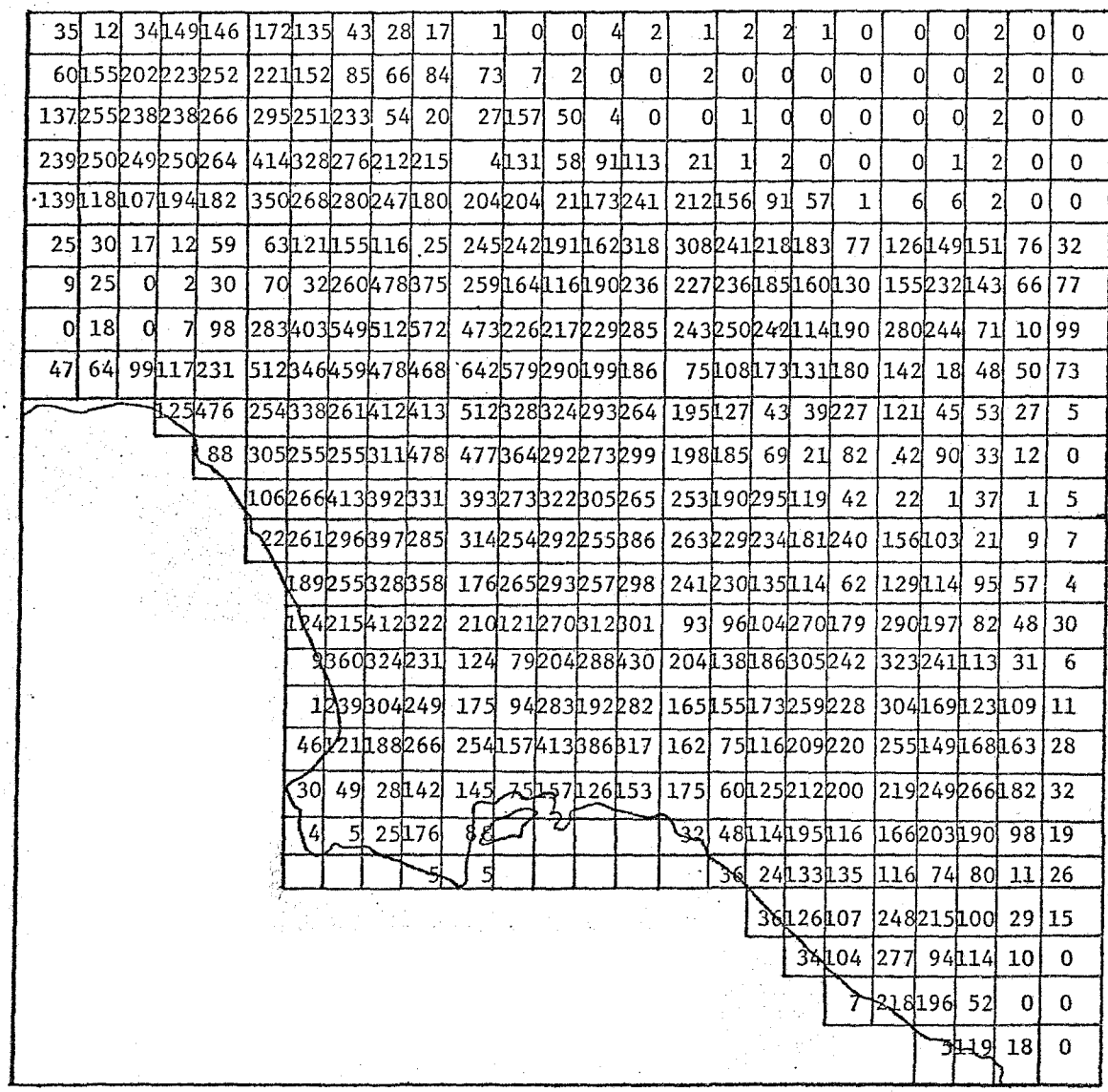

Figure 3 The Spatial Distribution of Surface Street Traffic, 1969 (thousands of vehicle miles traveled daily within each grid square).

in order to reflect the ratio of gasoline consumption to total highway fuel consumption for 1969 (California Statistical Abstract, 1970).

Emission factors for suspendible lead aerosol from automobiles can be calculated given the lead content of gasoline, vehicle fuel economy, and a detailed analysis of the fate of lead in gasoline. Data on the lead content of regular and premium priced gasolines marketed in Southern California during 1969 are given in Table 1 (Blade, 1969; Blade and Shelton, 1970). According to the Ethyl Corporation (1975), 60 percent of the gasoline sold in California during calendar year 1969 was premium grade, while the remainder was of lower octane. Assume that the average of the summer and winter lead contents of premium and regular gasolines is representative of that year's sales and that the statewide ratio of regular to premium grade sales is applicable to Los Angeles. The weighted average lead content of the 1969 Los Angeles area gasoline pool is then estimated to be 2.39 grams per gallon. Apparent fleet average fuel consumption in the Los Angeles Basin for 1969 is estimated to be about 12.9 miles per gallon (see Appendix 1). Other fuel economy estimates in use by public agencies range from 12 to 13.6 miles per gallon. Thus total 1969 Los Angeles lead consumption was 0.185 grams per vehicle mile traveled by gasoline fueled vehicles.

However, only a fraction of total automotive lead consumed contributes significantly to the lead burden of the atmosphere. Huntzicker, et al. (1975) have studied the fate of lead in the Los Angeles Basin. Their results are

Table 1

Lead Content of Gasoline

Southern California - 1969

	Lead Content in grams/gallon	
	Regular Grade	Premium Grade
Winter 1968-69	1.78	2.52
Summer 1969	2.03	2.92
AVERAGE	1.905	2.72

summarized in Figure 4. Of the tetraethyl lead initially present in gasoline, roughly 24.4 percent was estimated to be retained in the vehicle's oil, oil filter, and exhaust system. The remaining 75.6 percent of the lead consumed (i.e., 0.140 gm/mile for 1969) is emitted to the atmosphere. Ninety-three percent of these atmospheric emissions are in the form of a lead containing aerosol (about 0.130 gm/mile). The remainder is emitted as an organic vapor.

Particle size distribution plays an important part in determining atmospheric lead aerosol concentrations. Based on work by Habibi (1973), Huntzicker, et al. (1975) estimated that only 43 percent of the lead aerosol mass emitted at the tailpipe from typical Los Angeles traffic is in size ranges below 9μ (aerodynamic diameter) which can remain suspended at large distances from their source. The remaining 57 percent of the lead aerosol is in aerodynamically large particles which undergo rapid gravitational settling within a few hundred meters of the roadway.

The spatial distribution of annual average suspended lead concentrations over the Los Angeles Basin for 1969 is calculated using the advective Gaussian area source emissions to air quality model for a conserved pollutant proposed by Gifford and Hanna (1970). The computer program used for this air quality model is reproduced by Hanna (1972). For modeling purposes, only suspendible aerosol lead emitted at a rate of 0.056 gm/mile in 1969 will be considered to contribute to area-wide lead air quality (as measured by Hi Volume samplers),

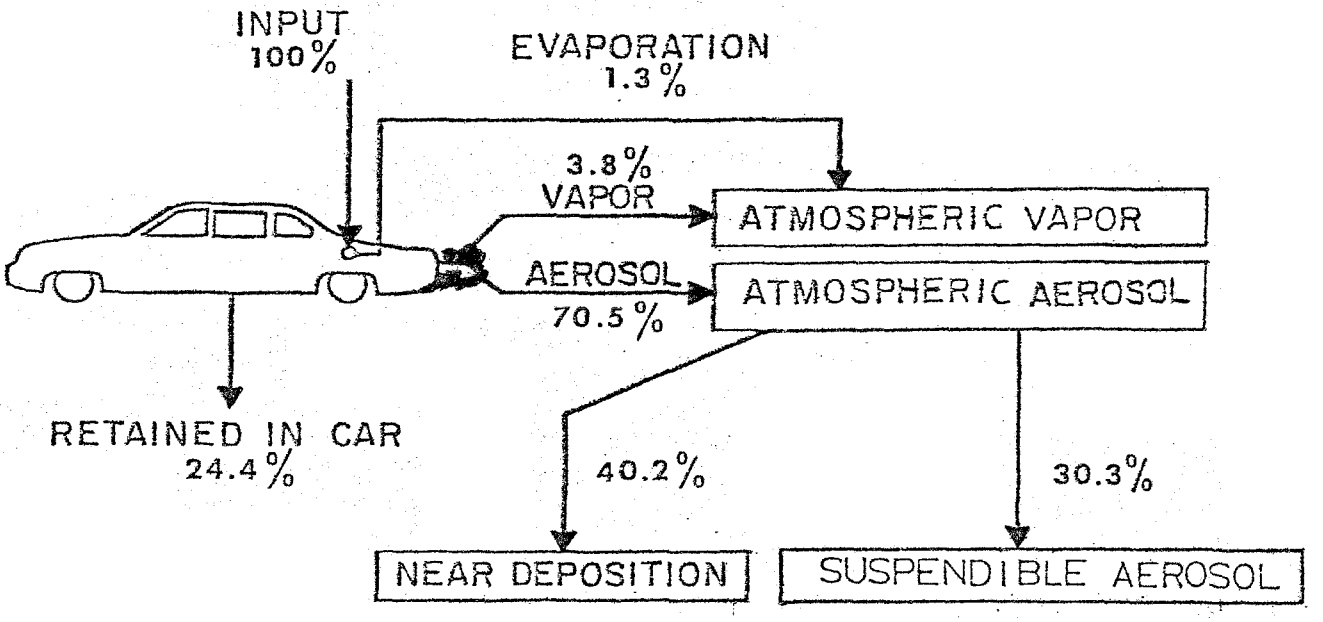

Figure 4 The Fate of Automotive Emitted Lead in the Los Angeles Basin (abstracted from Huntzicker, et al., 1975). All values shown as percent of lead initially present in gasoline.

with the understanding that the model is not expected to apply to lead measurements made immediately adjacent to a major freeway where near deposition has not yet depleted the aerosol lead in the larger particle sizes.

Wind speed and direction measurements taken by the Los Angeles Air Pollution Control District at downtown Los Angeles will be used in the dispersion calculations. The downtown Los Angeles meteorological station was chosen for use in this single wind rose air quality model because of its central location in the heart of the area of highest vehicle traffic density, and because it represents a compromise between the somewhat higher wind speeds recorded at coastal locations versus the generally lower wind speeds prevailing in the inland valleys to the northeast and northwest of downtown Los Angeles. The 1969 annual average wind speed at downtown Los Angeles is given as 2.37 meters per second (5.3 miles per hour; Los Angeles APCD, 1969). While the frequency distribution of wind directions at downtown Los Angeles is not readily available for 1969, such data has been compiled for the longer period 1956 through 1973 at that location, as shown in Table 2 (L.A. APCD, 1975a), and will be assumed to apply to 1969 in these calculations. In accordance with Gifford and Hanna's instructions for long term average air quality computations, using their model, neutral atmospheric stability will be assumed ($\sigma_z = 0.15X^{0.75}$; see Gifford and Hanna, 1970).

Table 2

Frequency of Wind from Direction Indicated

Downtown Los Angeles - 1956 through 1973

Direction	N	NNE	NE	ENE	E	ESE	SE	SSE	S	SSW	SW	WSW	W	WNW	NW	NNW
Relative Frequency	.054	.056	.129	.019	.073	.021	.056	.020	.108	.062	.233	.037	.096	.004	.020	.009

Annual average suspended lead levels computed by the above procedure for the 1969 traffic data are pictured in Figure 5. Contours of equal lead concentration were drawn at 1 microgram per cubic meter intervals on a Calcomp plotter using the Caltech system subroutine TOPOG. 1969 annual average suspended lead concentrations measured at nine locations by the National Air Sampling Network (EPA, 1973) and the Los Angeles Air Pollution Control District (MacPhee and Wadley, 1972) are indicated on the map. Substantially elevated lead measurements taken by the L.A. APCD at their Lennox air monitoring station are not included, since that station is located immediately adjacent to a busy freeway where large settleable lead particles would be expected to contribute significantly to measured values. Otherwise, agreement between calculated and measured lead concentrations is good considering the simplifications employed. Having gained some confidence that the automotive-related lead concentrations calculated by this method are in reasonable agreement with observation, we will turn to the question of projecting the long term average air quality impact of primary sulfuric acid mist emissions from catalyst-equipped cars.

Projecting the Impact of Direct Sulfuric Acid Mist Emissions from Catalyst-Equipped Cars

Data on the particulate emission characteristics of catalyst-equipped cars are relatively scarce at present. This discussion will be based on preliminary information gleaned from a U.S. Environmental Protection Agency

ANNUAL AVERAGE LEAD CONCENTRATION - 1969

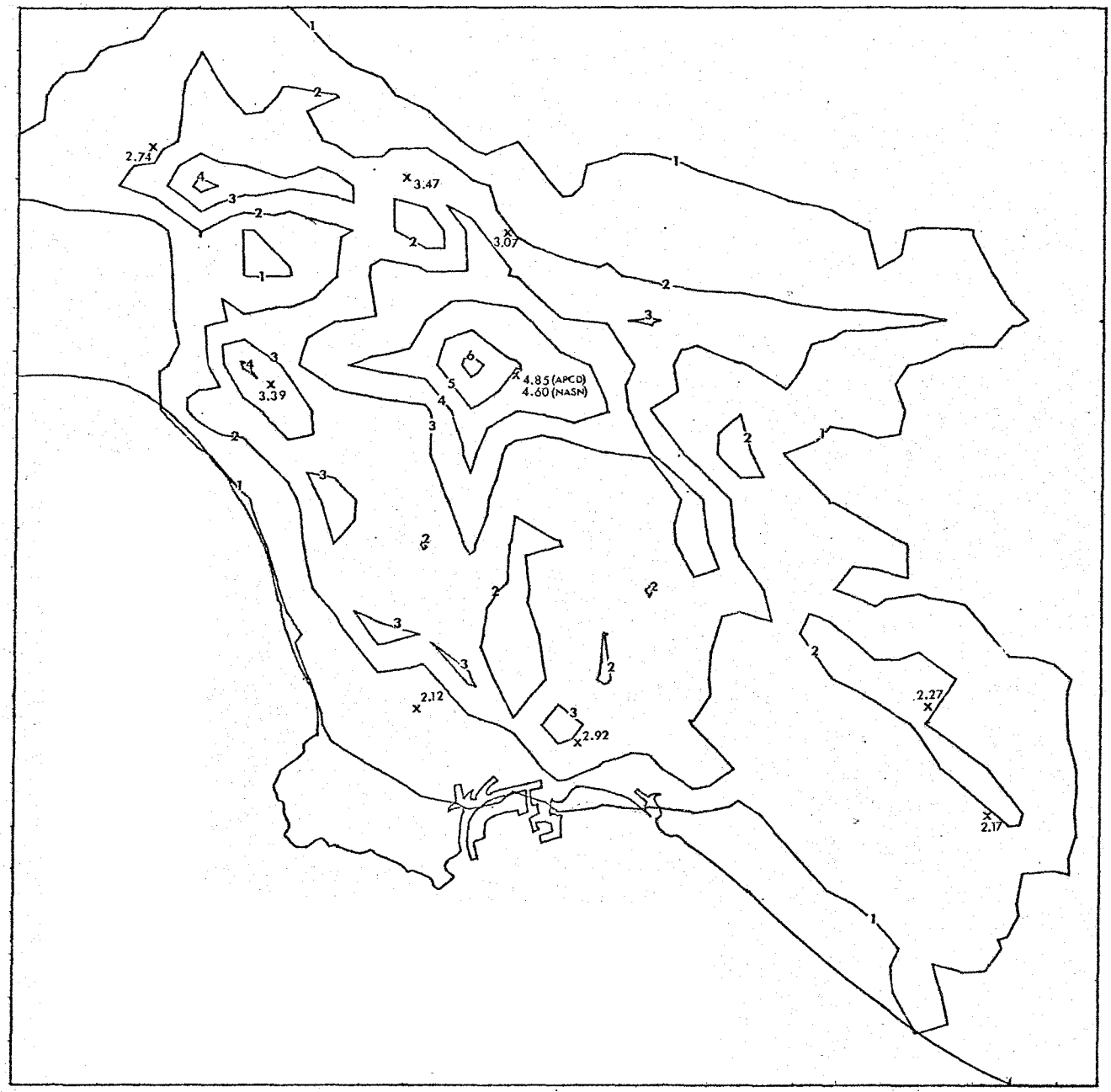

Figure 5 Annual Average Suspended Lead Concentrations for 1969 in the Los Angeles Basin. Isopleths are computed from traffic data. 1969 annual average measured lead concentrations are shown to two decimal places adjacent to monitoring station locations marked by X's. All values are in micrograms per cubic meter.

Issue Paper on the subject (EPA, 1975), and from a response to the EPA estimates prepared for the Manufacturers of Emission Controls Association by Energy and Environmental Analysis, Inc. (EEA, 1975).

From these two sources, it appears to be generally conceded that the catalyst emission control systems used on the 1975 model cars are capable of oxidizing a substantial portion of the sulfur contained in gasoline to form sulfuric acid mist at the vehicles' tailpipe. The actual rate of sulfuric acid mist emission depends at least upon the sulfur content of the fuel, the vehicle driving cycle, the type of catalyst, vehicle fuel economy, catalyst deterioration rate, and whether or not excess air is supplied to the catalyst (use of an air pump).

Possible permutations and combinations of future emission control systems, gasoline compositions, and vehicle use patterns are nearly endless. For that reason, a few illustrative examples will be worked to estimate the sulfate air quality impact of direct sulfuric acid mist emissions from catalyst-equipped cars under certain stated circumstances. *The objective is not to predict that these circumstances will come to pass, but rather to illustrate the methodology involved.* Since the form of industry and governmental response to the sulfuric acid mist emission controversy is not yet clear, examples will be chosen which approximate the air quality impact expected if trends apparent in 1975 model California cars were to be continued into future model years. Two cases will be considered. In the

first example, about three model years of catalyst-equipped cars are placed on the road and fueled exclusively with a selectively blended low sulfur unleaded gasoline. The second example will project the conversion of nearly the entire vehicle fleet to vehicles of the 1975 model type fueled by gasoline of a sulfur content equal to that of the entire current Southern California gasoline pool.

With all other factors held constant, it has been observed that sulfuric acid emissions from catalyst-equipped cars increase with increasing sulfur content of the fuel. Data presented in the Energy and Environmental Analysts report (EEA, Appendix 3, Figure 2) indicate that these increased emissions are linear with respect to gasoline sulfur content. The sulfur content of gasolines used in Southern California in the winter of 1973-74 is presented in Table 3 from Shelton (1974), along with the fraction of the 1974 California gasoline sales attributed to each grade of gasoline by Ethyl Corporation (1975). Weighting each fuel grade in proportion to its consumption, one estimates the sulfur content of the entire 1974 Southern California gasoline pool to be about 0.051 percent by weight. By selective blending, however, the sulfur content of the unleaded gasoline available to catalyst-equipped cars was at that time kept at about 0.026 percent, substantially below the gasoline pool average.

Driving cycle changes can have a dramatic effect on sulfuric acid mist emission rates. During cold-start, low-speed travel representative of an

Table 3

Sulfur Content of Gasoline

Grade	Southern California Gasoline Sulfur Content Winter - 1974 [Weight Percent]	% of 1974 total California sales for that grade
Premium	.039	43.5
Non-Premium		
Regular	.061	56.5
Unleaded	.026	(assumed small in 1974)

Table 4

Sulfuric Acid Emission Factors for 1975

Model Light Duty Motor Vehicles

HC/CO Emission Standard Met	Vehicle Model Year	Fuel Sulfur Content	Emission Factor Grams/Mile Urban Cycle	Highway Test
California Interim	1975	0.050%	0.025	0.08
California Interim	1975	0.051%	0.026	0.082
California Interim	1975	0.026%	0.013	0.042

urban driving cycle, catalyst systems, particularly those of the pelleted variety, tend to store sulfates on the surface of the catalyst beds. When the vehicle is brought to freeway speeds and catalyst temperatures increase, the stored sulfates are released as sulfuric acid mist. To deal with this change in emission rate, some combination of urban cycle and highway driving modes must be assumed. The approach taken here will be to use emission factors representative of the EPA's 1975 urban cycle test procedure (75 FTP) for surface street traffic, and the EPA 1975 Highway Fuel Economy Test Procedure cycle (HFET) as representative of freeway traffic.

Emission factors specific to automobiles manufactured to meet California Interim 1975 emission standards (HC: 0.9 gm/mi; CO: 9 gm/mi) are given by EPA (Issue Paper, 1975, Table 3). These emission factors were based on a fuel sulfur content of 0.05 percent, the use of a fleet mix of catalyst emission control system types (40 percent pelleted, 60 percent monolithic), and the assumption that vehicles will be equipped with excess air injection in order to meet the California emission standards for exhaust hydrocarbons and carbon monoxide. In Table 4, these emission factors have been adjusted to the basis of 0.051 percent sulfur and 0.026 percent sulfur, using the assumption of emission linearity with respect to sulfur content. This was done in order to better reflect the two cases of interest here: (1) use of current unleaded fuel with 0.026 percent sulfur, and (2) possible future existence of an unleaded gasoline pool reflecting the entire current 0.051 percent sulfur content of Southern California gasolines.

As was the case with automotive lead emissions, particle size must be considered when estimating the air quality impact of sulfuric acid mist emissions. In this case, however, particle deposition rates would be expected to be very different. Sulfuric acid mist generally nucleates to form a fine submicron aerosol which is unlikely to undergo significant gravitational settling within a scale of tens of kilometers from the source. Actual particle size distribution measurements specific to sulfuric acid mist from catalyst-equipped cars are scarce in the literature. However, total particulate emission size data taken by Moran, et al. (1972, run #41) from a catalyst-equipped vehicle whose exhaust was identified as containing a high fraction of sulfur-bearing aerosols indicated that 97 percent of the total particulate mass distribution from that test was in sizes less than one micron (aerodynamic diameter). Thus it will be assumed that atmospheric fractionation by gravitational settling will be insignificant, and that sulfate ion emissions from catalyst-equipped cars can be treated as a conserved species for area source modeling purposes.

Vehicle use patterns are a further important determinant of the future impact of catalyst technology. In addition to the driving cycle considerations mentioned, one must be able to project the future spatial distribution of vehicle traffic and the rate of introduction of new cars into the fleet. New car purchases and vehicle miles traveled are both sensitive to current economic conditions. Accurate extrapolation of historic trends into the

future is quite difficult. However, some assumption must be made in this regard. For the purposes of these examples, the rate of introduction of new cars into the Los Angeles vehicle fleet will be taken from historic trends as given by Lees, et al. (1972) and shown in Table 5. The spatial distribution of vehicle miles traveled will be estimated by using the given 1969 traffic patterns as a baseline, adjusting as before for the ratio of gasoline to total highway fuel consumption, and then increasing traffic volume to the desired future year using a spatially uniform compound annual growth rate. Since this approximation is unlikely to be strictly correct, both a high growth rate case (4 percent per year) and a low growth rate case (2 percent per year) will be shown for each example of interest.

The emissions to air quality model previously validated for lead particulates is next applied to the direct emission of sulfuric acid mist from catalyst-equipped cars. A tabulation of the assumptions involved in each illustration is given in Table 6. In keeping with the long term annual average air quality levels sought, the 1956-1973 wind rose for downtown Los Angeles is again used, and this time the 2.42 meter per second 1956-1973 average wind speed at downtown Los Angeles is also employed (L.A. APCD, 1975a).

Results of the example with three model years of catalyst-equipped cars and lower sulfur fuel are shown in Figures 6 and 7 for low and high growth rate cases respectively. Results of the example illustrating nearly complete fleet conversion and a higher sulfur fuel are shown in Figures 8 and 9 for low and high growth rate cases respectively.

Table 5

Birth-Death Schedule for

Los Angeles Area Automobiles

Vehicle Age (Years)	% of Vehicle Population	Miles Driven Yearly	% of Vehicle Miles Traveled By Cars of that Model Year
0	11	15,000	19.5
1	11	13,000	16.9
2	11	11,000	14.3
3	10	9,600	11.3
4	10	8,400	9.9
5	9	7,600	8.1
6	8	5,300	5.0
7	7	5,000	4.1
8	6	4,400	3.1
9	5	4,200	2.5
10	4	3,500	1.7
11	3	3,500	1.2
12	2	3,500	0.8
13	2	3,500	0.8
14	1	3,500	0.4
15	1	3,500	0.4

Table 6

Summary of Assumptions for Sulfate Air Quality Computations

Case	Fraction of Gasoline-Powered Vehicle Miles Traveled with Oxidation Catalysts	Approximate Number of Model Years Involved	Approximate Year	Sulfur Content of Gasoline	$SO_4^=$ Emission Factors (gm/mile) Urban Cycle	$SO_4^=$ Emission Factors (gm/mile) Highway Test	Traffic Growth Multiplier Applied to 1969 Traffic Counts
IA	50.7%	3	1978	0.026%	0.013	0.042	9 yr @ +2%/yr = 1.195
IB	50.7%	3	1978	0.026%	0.013	0.042	9 yr @ +4%/yr = 1.423
IIA	94.7%	10	1985	0.051%	0.026	0.082	16 yr @ +2%/yr = 1.373
IIB	94.7%	10	1985	0.051%	0.026	0.082	16 yr @ +4%/yr = 1.873

ANNUAL AVERAGE SULFATE LEVELS FROM CATALYST EQUIPPED AUTOS - CASE I A

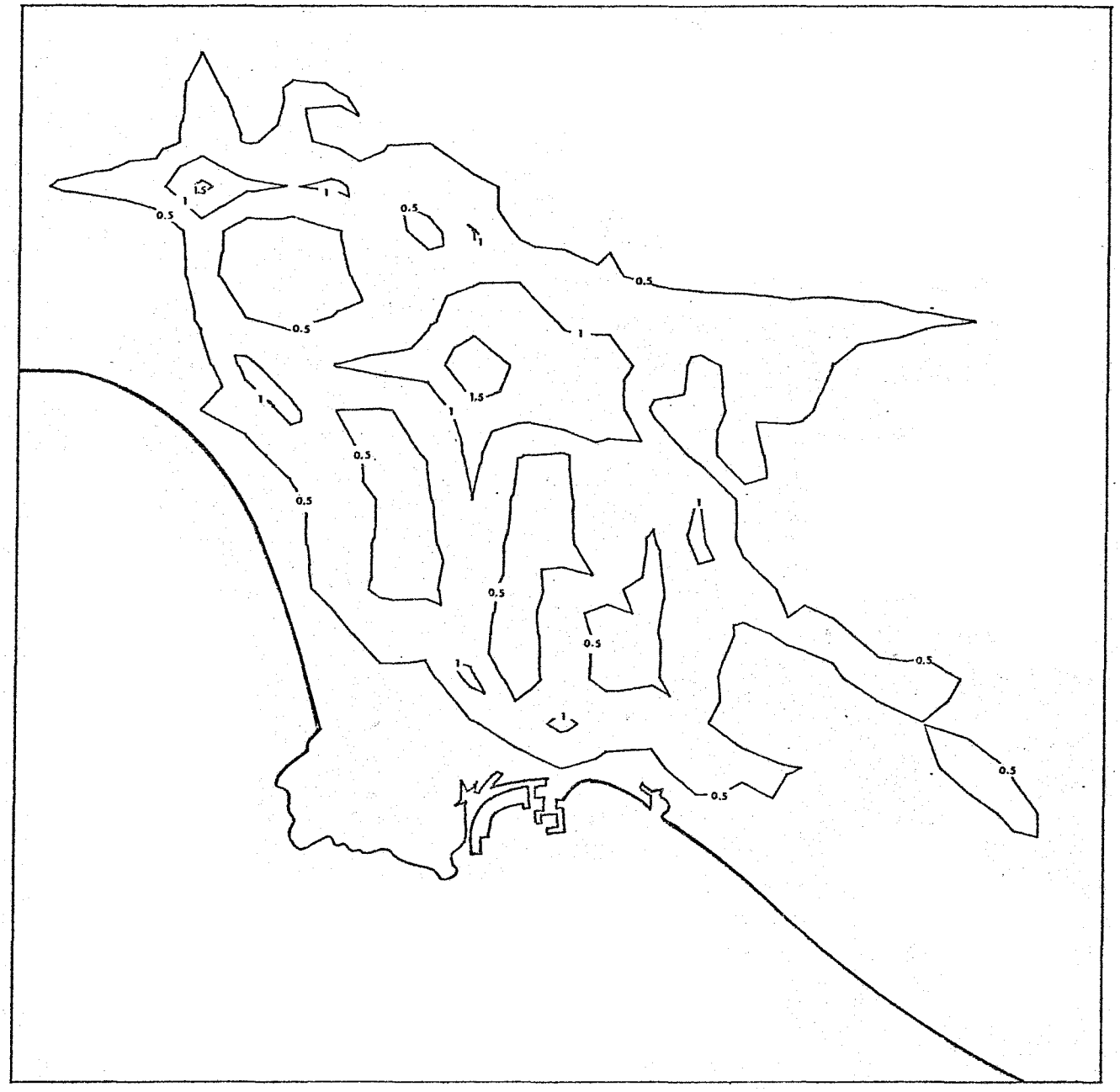

Figure 6 Projected Annual Average Sulfate Levels from Catalyst-Equipped Autos - Case IA. All values in $\mu gm/m^3$ as H_2SO_4. (See Table 5.6 for list of assumptions.)

ANNUAL AVERAGE SULFATE LEVELS FROM CATALYST EQUIPPED AUTOS - CASE I B

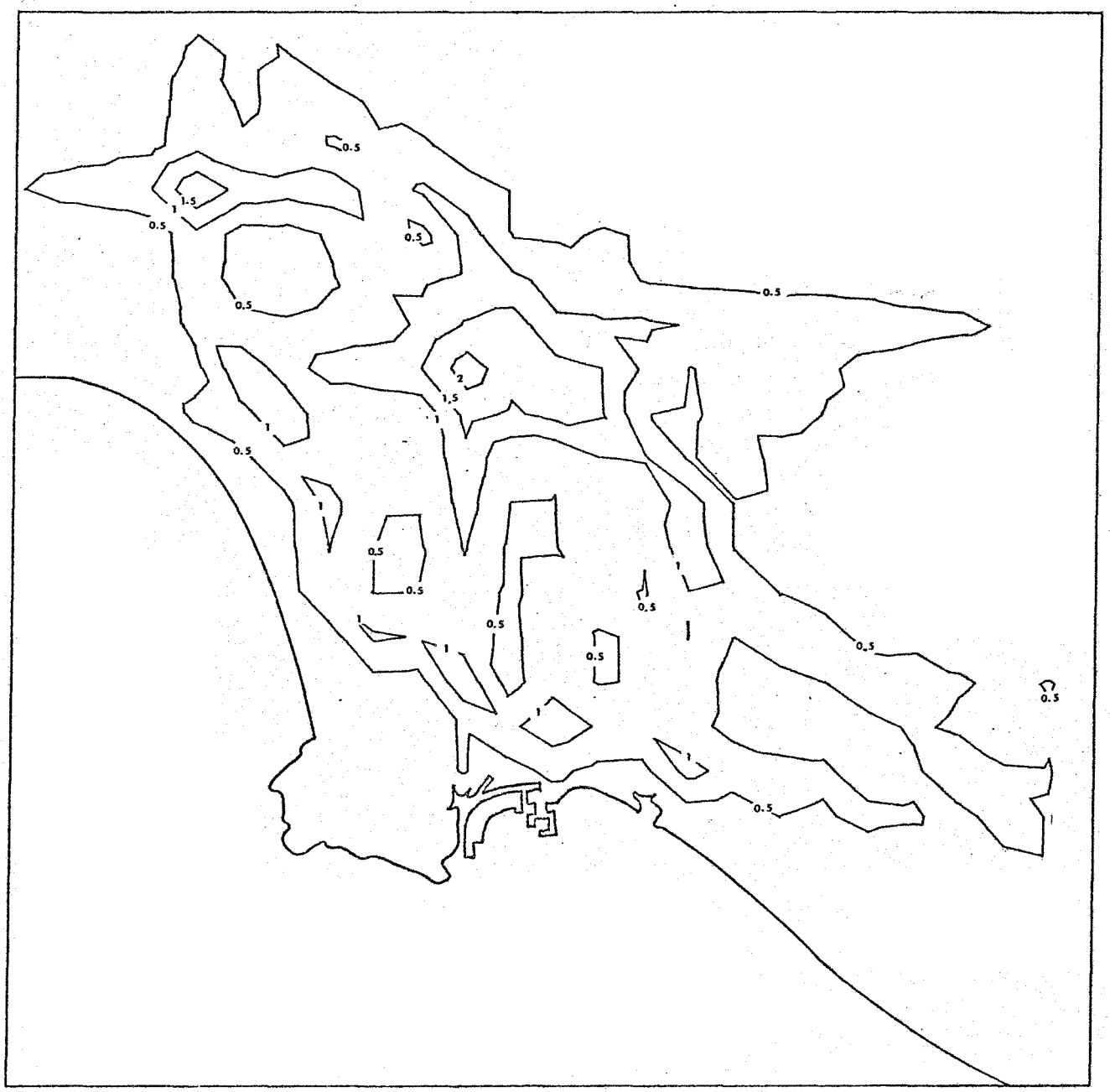

Figure 7 Projected Annual Average Sulfate Levels from Catalyst-Equipped Autos - Case IB. All values in $\mu gm/m^3$ as H_2SO_4. (See Table 5.6 for list of assumptions.)

ANNUAL AVERAGE SULFATE LEVELS FROM CATALYST EQUIPPED AUTOS - CASE II A

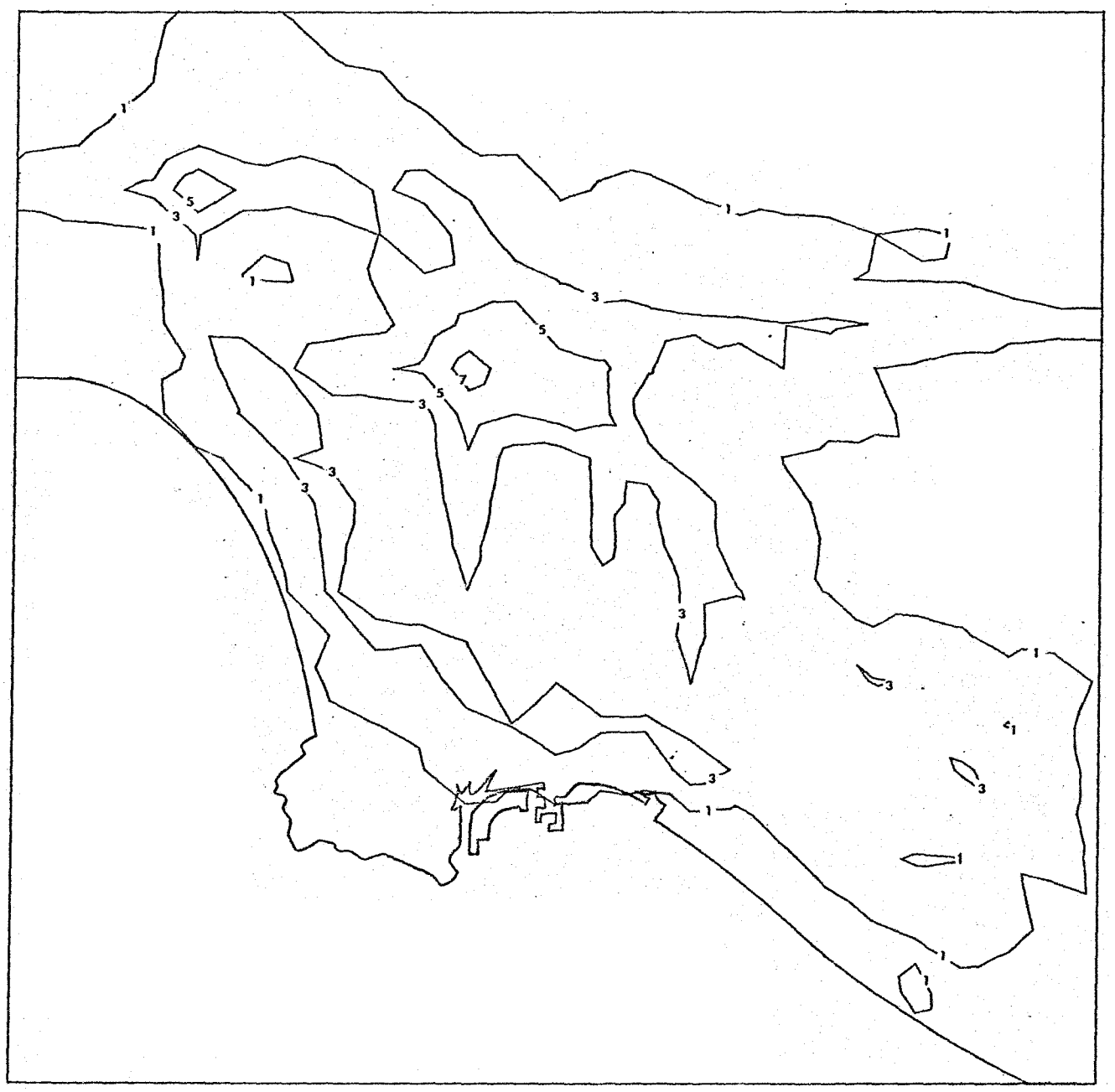

Figure 8 Projected Annual Average Sulfate Levels from Catalyst-Equipped Autos - Case IIA. All values in $\mu gm/m^3$ as H_2SO_4. (See Table 5.6 for list of assumptions.)

ANNUAL AVERAGE SULFATE LEVELS FROM CATALYST EQUIPPED AUTOS - CASE II B

Figure 9 Projected Annual Average Sulfate Levels from Catalyst-Equipped Autos - Case IIB. All values in $\mu gm/m^3$ as H_2SO_4. (See Table 5.6 for list of assumptions.)

Discussion

The air quality model demonstrated in Cases I and II was applied only to the incremental impact of direct emission of sulfuric acid mist from catalyst-equipped cars. In order to assess the significance of this emerging automotive pollution problem, one must consider the pre-existing Los Angeles Basin sulfate air quality levels with which the effluent from catalyst-equipped cars will be mixed. Over the past half decade, Los Angeles area annual average sulfate air quality levels have generally ranged between about 10 and 15 micrograms per cubic meter. The spatial distribution of arithmetic average sulfate air quality for the period July 1971 through June 1973 is shown in Figure 10 (TRW, 1974).

The concentration patterns arising from direct sulfuric acid mist emissions from catalyst-equipped cars cannot be blindly superimposed upon existing sulfate air quality levels. The pre-existing sulfate concentrations are largely due to a combination of natural sulfate sources plus sulfates produced by atmospheric reactions involving sulfur dioxide gas emitted from certain industrial processes and from burning sulfur-bearing fuel (TRW, 1974). In 1974, automobiles contributed about 30 tons per day of the 365 tons per day of sulfur dioxide emissions in Los Angeles County (L.A. APCD, 1975b). Direct conversion of a portion of the sulfur in gasoline to sulfuric acid mist will somewhat reduce these automotive sulfur dioxide emissions. However, as an order of magnitude approximation, the set of circumstances postulated in

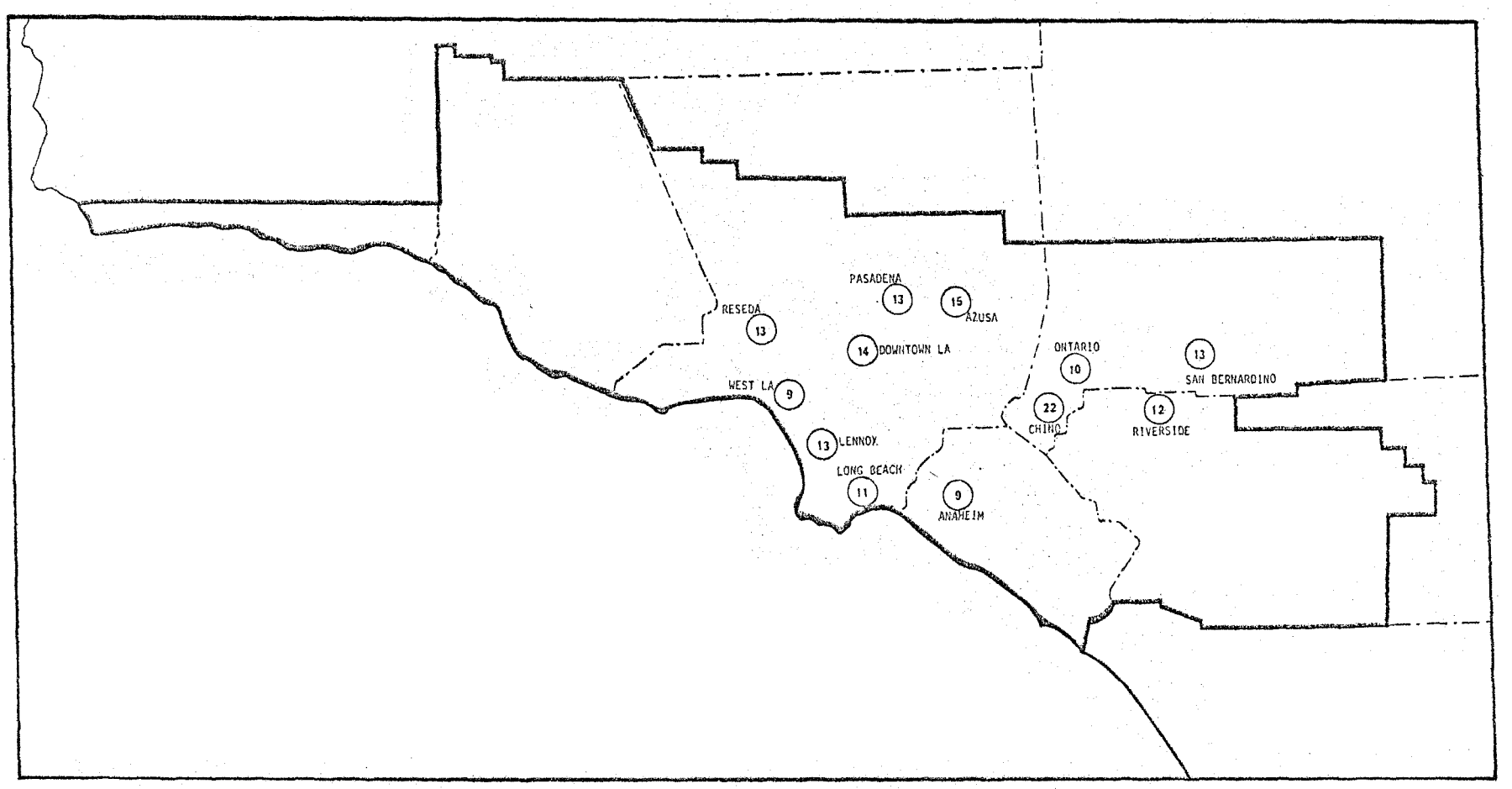

Figure 10 The Spatial Distribution of Arithmetic Average Sulfate Air Quality in the Los Angeles Area - July 1971 through June 1973 (from TRW, 1974). All values in $\mu gm/m^3$ as $SO_4^=$.

Case II (complete conversion of the fleet to 1975-type cars, combined with no reduction in total sulfur in the gasoline pool) could increase annual average sulfate levels in portions of downtown Los Angeles by roughly two-thirds by 1985 if no changes take place to alter 1975 vehicle designs or to lower the sulfur content of the gasoline pool. On the other hand, as indicated in Case I, a few model years of 1975-type catalyst-equipped cars will not drastically affect annual average sulfate levels over wide areas of the Basin provided that the sulfur content of unleaded fuel is kept relatively low through selective blending.

Acknowledgment

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APPENDIX 1

Gasoline Powered Vehicle Fuel Economy

Gasoline consumption will be estimated as follows:

$$E_t = E_a \gamma + E_o (1 - \gamma)$$

Where

E_t = fleet average fuel consumption by gasoline powered vehicles, 1969

E_a = fleet average automobile fuel economy, 1969

E_o = fleet average fuel economy for non-automotive gasoline vehicles, 1969

γ = fraction of gasoline-fueled vehicle miles traveled in modeling region by automobiles

All data will be taken from Reference (1) below.

E_a = 13.63 miles/gallon for U.S. auto fleet in 1969

E_o = 8.36 miles/gallon for U.S. truck fleet in 1969 (assumed equal to gasoline-fueled non-auto traffic fuel economy for that year)

As was assumed by Roth, et al. (1974), vehicle miles traveled within this modeling region will be taken as proportional to vehicle registrations.

$$\gamma \approx \frac{R_a}{R_a + R_o (1 - \Delta)}$$

R_a = Los Angeles County auto registration as of 7/1/70 = 3,270,123

R_o = Los Angeles County truck registration as of 7/1/70 = 537,375

$$\Delta = \frac{\text{Calif. diesel truck \& bus registration, 1969}}{\text{Total Calif. truck \& bus registration, 1969}}$$

$$\Delta = \frac{61,788 + 7,459}{1,915,178} = 0.036$$

$$\gamma = \frac{3,270,123}{3,270,123 + (537,375)\ (0.964)} = 0.86$$

Then $E_t = (13.63)\ (0.86) + 8.36\ (0.14)$

E_t = 12.9 miles/gallon

References: (1) Pekkala, Peter E., "1971 Automobile Facts and Figures," Detroit, Automobile Manufacturers Association, Inc., 1971.